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**SURFACE PHOTOVOLTAGE SPECTROSCOPY APPLIED TO  
GALLIUM ARSENIDE SURFACES**

by  
**Charles E. Byvik**

**December 1975**

**(NASA-TM-X-71958) SURFACE PHOTOVOLTAGE  
SPECTROSCOPY APPLIED TO GALLIUM ARSENIDE  
SURFACES Interim Release (NASA) 23 p HC  
\$3.50**

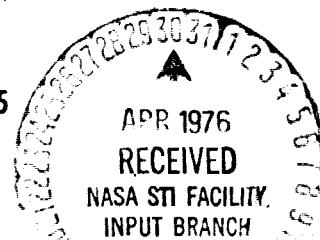
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LANGLEY RESEARCH CENTER, HAMPTON, VIRGINIA 23665**



1. Report No. <b>TMX-71958</b>		2. Government Accession No.		3. Recipient's Catalog No.	
4. Title and Subtitle <b>Surface Photovoltage Spectroscopy Applied to Gallium Arsenide Surfaces</b>				5. Report Date <b>December 1975</b>	
				6. Performing Organization Code	
7. Author(s) <b>Charles E. Byvik</b>				8. Performing Organization Report No.	
				10. Work Unit No.	
9. Performing Organization Name and Address <b>Langley Research Center National Aeronautics and Space Administration Hampton, VA 23665</b>				11. Contract or Grant No.	
				13. Type of Report and Period Covered <b>Technical Memorandum</b>	
12. Sponsoring Agency Name and Address <b>National Aeronautics and Space Administration Washington, DC 20546</b>				14. Sponsoring Agency Code	
15. Supplementary Notes <b>Interim technical information release, subject to possible revision and/or later formal publication.</b>					
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17. Key Words (Suggested by Author(s)) <b>Gallium Arsenide Surface States Surface Voltage</b>				18. Distribution Statement	
19. Security Classif. (of this report) <b>Unclassified</b>		20. Security Classif. (of this page) <b>Unclassified</b>		21. No. of Pages <b>20</b>	
				22. Price <b>\$3.25</b>	

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SURFACE PHOTOVOLTAGE SPECTROSCOPY APPLIED  
TO GALLIUM ARSENIDE SURFACESby  
Charles E. Byvik

## I. SUMMARY

The experimental and theoretical basis for surface photovoltage spectroscopy is outlined. Results of this technique applied to gallium arsenide surfaces, both reported in the literature and obtained at Langley Research Center, are reviewed and discussed. The results suggest that in GaAs the surface voltage may be due to deep bulk impurity acceptor states that are pinned at the Fermi Level at the surface. Establishment of the validity of this model will indicate the direction to proceed to increase the efficiency of GaAs solar cells.

## II. INTRODUCTION

The mechanism responsible for limiting the efficiency of gallium arsenide (GaAs) solar cells is considered to be the high surface recombination velocity (SRV) (ref. 1). Lowering the SRV will result in a more efficient solar cell. The SRV is a parameter defined as the ratio of the rate of carrier flow into a unit surface area to the excess carrier density in the bulk just beneath the surface. Using a model that is essentially the Shockley-Read bulk recombination model extended to discrete surface states, one finds (ref. 2) that the SRV depends on the capture cross sections of the surface states for electrons and holes, the density of surface states, the doping density of the bulk, the energy position of the surface states, the temperature, and the surface voltage. A technique that has been utilized to experimentally measure these parameters in wide bandgap semiconductors (SC) utilizes the

Kelvin probe and sub-bandgap monochromatic radiation, and is called (refs. 3-15) surface photovoltage spectroscopy (SPS). It is the intent of this report to briefly review the theory of SPS as it applies to the wide bandgap SC material gallium arsenide, discuss the experimental set-up at Langley Research Center, report the results to date obtained by this technique, and discuss the implications of these results.

### III. SYMBOLS

$E_c$	energy of the conduction band edge
$E_F$	Fermi energy
$E_i$	intrinsic energy
$E_t$	trap energy
$E_v$	energy of the valence band edge
$f(E)$	Fermi function
$F_s$	space charge function at the surface
$h$	Planck's constant
$I$	radiation intensity
$K_n$	capture cross section for electrons multiplied by their thermal velocity
$K_p$	capture cross section for holes multiplied by their thermal velocity
$K_{ph}^d$	capture cross section for photon-induced electron transitions from surface states to the bulk
$K_{ph}^p$	capture cross section for photon-induced electron transitions from the bulk into the surface states
$k$	Boltzmann's constant
$L$	effective Debye length
$N_t$	surface density of trap states
$n_B$	bulk electron density
$n_i$	intrinsic electron density
$n_s$	surface density of free electrons
$n_s^0$	equilibrium surface state density of electrons under dark conditions
$n_t$	electron density in the surface states
$n_l$	emission coefficient
$p_t$	$= N_t - n_t$

$p_t^0$	$= N_t - n_t^0$
$Q_{sc}$	space charge density
$Q_{ss}$	surface charge density
$q$	electronic charge
$S$	surface recombination velocity
$S_{max}$	maximum value of $S$
$T$	absolute temperature
$t$	time
$U_0$	$= 1/2 \ln(K_p/K_n)$
$U_s$	surface potential in the dark in $kT/q$ units
$V$	electron potential
$V_s$	surface voltage
$v$	$= qV/kT$
$W_R$	reference work function
$W_s$	sample work function
$z$	spatial position
$\epsilon$	dielectric permittivity
$\nu$	frequency
$\rho$	charge density
$\chi$	electron affinity

Abbreviations:

CPD	contact potential difference
SRV	surface recombination velocity

#### IV. THEORY

The theory of surface photovoltage spectroscopy (SPS) reviewed in this section will be from a phenomenological viewpoint. If a conductor is placed near the surface and the capacitance of the SC-conductor system is monitored, then any change in occupation of the surface states will result in a capacitance change. The basis for the success of SPS is the detection of a change in capacitance due to the depopulation of the surface states by monochromatic light having a wavelength corresponding to the energy difference between the

surface states and the conduction band. Therefore, by sweeping monochromatic radiation with sub-bandgap energy and by monitoring capacitance, the energy positions of the surface states can in principle be determined.

For definiteness, an n-type SC having acceptor states located on the surface at a discrete energy  $E_t$  and density  $N_t$  will be considered. Under dark conditions, at a temperature  $T$ , these states will be occupied with electrons to the density  $n_t$  where

$$n_t = N_t f(E_t)$$

where  $f(E_t)$  is the Fermi-Dirac distribution function. Since these states are localized at the surface and are occupied by electrons, a region near the surface will become depleted of the mobile conduction electrons which will result in a space charge region consisting of the immobile, positively ionized donors. This dipole layer gives rise to a surface voltage extending into the material a distance  $L$ , called the effective Debye length (ref. 2). See figure 1.

The equations pertinent to SPS are Poisson's equation and the kinetic equations

$$\nabla^2 v = - \frac{4\pi q}{\epsilon kT} \rho \quad (1)$$

$$\frac{dn_t}{dt} = (K_{ph}^p p_t - K_{ph}^d n_t) I + K_n (n_s p_t - n_t n_l) \quad (2)$$

and in the dark at thermal equilibrium

$$n_s^o p_t^o - n_t^o n_l = 0 \quad (3)$$

where the potential  $v$  is written in units of  $\frac{kT}{q}$ ,  $k$  is Boltzmann's constant,  $q$  is the electronic charge,  $\epsilon$  is the dielectric permittivity,  $p_t$  is the density of holes in the surface states,  $\rho$  is the charge density,  $n_s$  is the density of free electrons at the surface,  $n_l$  is the emission



coefficient,  $I$  is the light intensity,  $K_n$  is the capture cross section for electrons multiplied by thermal velocity, and  $K_{ph}^d$  and  $K_{ph}^p$  are the capture cross sections for photon-induced electron transitions involving surface state depopulation and population, respectively. The zero superscripts in equation (3) indicate thermal equilibrium and dark conditions.

These equations assume a single set of states at the surface. The case of multiple states (refs. 3-14) and the effects of surface states in good communication with the bulk on the detection of multiple states (ref. 10) are discussed in the literature.

The solution to Poisson's equation with the boundary conditions that at  $z = 0$ ,  $V = V_s$  and for  $z \gg L$ ,  $V = 0$  is

$$Q_{ss} = qn_t = -Q_{sc} = -q n_B L F_s \quad (4)$$

where  $Q_{ss}$  and  $Q_{sc}$  are the total charges in the surface states and space charge region, respectively,  $n_B$  is the carrier concentration in the bulk,  $L$  is the effective Debye length, and  $F_s$  is the space charge function at the surface (ref. 2). For a Schottky-type surface barrier equation (4) becomes

$$Q_{ss} = - (2 \epsilon q n_B)^{1/2} |V_s|^{1/2} = q n_t \quad (5)$$

The Schottky model indicates that the surface voltage,  $V_s$ , depends on the square of the number density of charges trapped in the surface states. This quadratic dependence enhances the sensitivity of the SPS technique.

Two competing processes are considered in equation (2), photon-electron interactions and phonon-electron interactions. It is assumed that the photon capture cross sections are step functions having the properties that

$$K_{ph}^d(\nu) \begin{cases} = 0 & \text{for } h\nu < E_c - E_t \\ \neq 0 & \text{for } h\nu \geq E_c - E_t \end{cases} \quad (6)$$

$$K_{ph}^p(\nu) \begin{cases} = 0 & \text{for } h\nu < E_t - E_v \\ \neq 0 & \text{for } h\nu \geq E_t - E_v \end{cases} \quad (7)$$

where  $E_v$  and  $E_c$  are the energies of the valence band and conduction band edges, respectively,  $h$  is Planck's constant and  $\nu$  is photon frequency. With a proper choice of experimental conditions, the following surface state parameters can be determined: photon capture cross section, the surface voltage, capture cross section for electrons, the density of states, and the fractional occupancy of states. (See reference 5 for the details.)

Experimentally, the surface photovoltage is measured as a change in the steady state contact potential difference (CPD) with respect to a vibrating gold reference electrode. The CPD is the difference in the work functions of the two materials, i.e.

$$CPD = W_R - W_S = W_R - (\chi_E + E_F - q V_S) \quad (8)$$

where  $W_R$  and  $W_S$  are the work functions for the reference electrode and the sample;  $\chi_E$  is the SC electron affinity;  $E_F$  is the Fermi Energy in the bulk; and  $V_S$  is the surface voltage. If there is any change in surface charge, then the CPD changes as a result of a change in  $V_S$  which can in turn be attributed to a change in the trapped surface charge through equation (4) or (5). It is the change in  $V_S$  that is monitored in SPS.

## V. EXPERIMENTAL PROCEDURE

A schematic of the experimental equipment used to monitor the CPD is shown in figure 2. A 150-watt xenon lamp is the light source which is collimated onto the entrance slit of a double prism monochromator. The radiation exiting from the monochromator is passed through a quartz window into a vacuum chamber and focused on the sample with a quartz lens. Care is taken to keep the intense part of the radiation from falling on the gold electrode and the electrical contacts on the sample.

A preamplifier is connected to the sample through a 10K ohm load resistor. Tin-silver ohmic contacts were made near the edge of the sample. The preamplifier operating in the vacuum amplified the signal by approximately  $10^4$  times and proved to be linear over the experimental operating range. The amplified signal was fed into a lock-in detector and the conditioned signal then recorded on a strip chart recorder. The reed was sized to have a resonant frequency of 90 Hz and with the gold probe mounted on the tip, resonated at approximately 115 Hz. The reed is driven by a coil carrying a signal from the signal generator. Care was taken to isolate the conductors carrying current to the driving coil from the signal leads.

The sample was etched using a solution of  $\text{1HF} + 3\text{HNO}_3 + 2\text{H}_2\text{O}$  for 30 seconds and then rinsed well in distilled water. The sample was isolated from ground and placed in an ion pumped vacuum system. The system was evacuated to approximately  $10^{-7}$  torr.

The sensitivity of the system was typically a few millivolts with a time constant of 3 seconds. This sensitivity depended on the magnitude of the surface voltage. The response of the system was checked using a cadmium sulfide sample which exhibited states corresponding to levels reported previously in the literature.

## VI. RESULTS

A photovoltage spectrum of the surface of n-type GaAs having a (111) orientation and taken from reference 11 is shown in figure 3. Also shown

in figure 3 is an insert which shows the derivative of the surface photovoltage with respect to energy. This procedure enhances the energy position of the surface states. From equations (8) and (5) and in the Schottky barrier approximation assuming only photodepopulation of the surface states,

$$\text{CPD} \propto V_s \propto n_t^2$$

Then

$$\left| \frac{\delta \text{CPD}}{\delta h\nu} \right| \propto \left| \frac{\delta V_s}{\delta h\nu} \right| \propto n_t^2 \left| \frac{\delta k_{ph}^d}{\delta h\nu} \right|$$

Since a step function behavior is assumed for photon interaction (equations (6) and (7)), an extremum will be exhibited at a photon energy corresponding to the trap energy. Similarly, an extremum will occur if photopopulation of the surface states occurs. The energy positions of these two extremums are related in that their sum is equal to the energy of the forbidden gap.

There are two maxima indicated in the insert that are attributed to a discrete surface state at 0.72 eV below the conduction band edge and a second level 0.92 eV below the conduction band edge. However, because the 0.72 eV state is in good communication with the bulk, recombination interferes with the determination of the position of this state. The 0.92 eV peak cannot be exactly located but lies at a position between 0.9 and 1.0 eV. Figure 4 is a schematic representation of the surface states indicated by this data.

Figure 5 is a photovoltage spectrum typical of the spectra for n-type GaAs (100) surfaces measured at Langley Research Center. It should be noted that there is no indication of surface states in the bandgap. None of the samples that were tested indicated any of the surface structure found in reference 11. Changes in the ambient gas conditions or surface treatment did not change the structure of the photovoltage curves but did change the magnitude of the surface voltage.

The resistivity of the sample as tested ranged from approximately 1 ohm-cm to  $10^{-3}$  ohm-cm. No structure in the mid-bandgap region was observed on any of these samples. Oxygen and water vapor ambients to pressures of 10 microns of Hg did not alter the photovoltage spectra although changes in the magnitude of the surface voltage were observed. The detailed behavior of the surface

voltage as a function of the ambient conditions was not pursued. The surface voltage, as determined by saturating the samples with intense white light and measuring the total change in CPD, ranged from 200 mV to 350 mV. This should be compared to the 550 mV surface voltage as measured by Lagowski, et al (ref. 11).

The lack of structure measured at Langley was confirmed by a comparison measurement made at MIT under the direction of Prof. H. C. Gatos.

## VII. DISCUSSION

A comparison between the results obtained in this laboratory with the data in the literature for GaAs leads to important questions regarding the origin of the surface states. Lagowski, et al (ref. 11) observed surface states at energies 0.72 eV and between 0.9 and 1.0 eV below the conduction band. These states were observed on a (111) oriented surface and by selective etching were found both on the (111) gallium exposed surface and on the (111) arsenic exposed surface. It should be noted that the SRV calculated using the Shockley-Read formulation (ref. 2) and the parameters measured for the 0.72 eV state results in SRV values typical of those observed for GaAs solar cells (ref. 15) (See Appendix.). However, since no states have been observed in this laboratory on the (100) oriented surfaces, two explanations are possible. One, because of the inherent nature of the (100) oriented surface, the density of surface states at these energies decreases to below the detectable limits of the experimental apparatus or the trap energy shifts or both. Two, the states observed are not intrinsic to the surface but are impurity states.

As for the first possibility, Valahas, et al (ref. 13) observed no significant changes in capture cross section or energy shifts in the trap level at 0.72 eV below the conduction band edge either on a (100) or a (111) oriented GaAs surface. Further, no significant changes in the surface state parameters were observed when the surface was treated with different etchants. These data lead to the conclusion that the surface state at 0.72 eV below the conduction band edge is not associated with a particular termination of the

lattice at the surface. Therefore, the surface states observed in the forbidden band may be associated with a deep impurity acceptor level in n-type GaAs which is pinned at the Fermi level, causing a depletion region and resulting in the photovoltage spectra observed. Ion probe mass spectrometer analysis of three different n-type GaAs samples indicates that there is sufficient density of bulk impurities, known to result in acceptor state levels in GaAs, to account for the density of surface states reported in the literature (ref. 11).

Gatos and Lagowski (ref. 9) argue that the majority carriers generated from deep impurity states cannot be the cause of the changes in surface photovoltage observed since theory shows a square root dependence for the surface voltage on the majority carrier concentration. A small change in surface voltage would therefore only be a result of a large change in photoconductivity; an effect not experimentally observed. However, the theory of surface voltage employed by Gatos and Lagowski assumes that the surface states are intrinsic to the surface of GaAs and that these states give rise to the observed surface voltage. If the assumption is made that these are not intrinsic surface states but deep impurity traps distributed homogeneously throughout the bulk and extending to the surface of GaAs and that these traps are acceptor levels in n-type GaAs (and a set of deep impurity levels that are donors in p-type GaAs), then the depletion layer and corresponding surface voltage can be explained in terms of demanding that these states be filled according to Fermi-Dirac statistics. The lack of photoconductivity observed by Gatos and Lagowski (ref. 9) may be due to rapid bulk recombination in the bulk.

Some in the literature support the impurity surface state model for GaAs. (1) Only depletion layers have been observed for GaAs. (2) Valahas, et al. (ref. 13) have observed a surface level at a position 0.45 eV below the conduction band in p-type GaAs doped with oxygen which they attribute to the bulk oxygen level. (3) The 0.72 eV level observed by Lagowski, et al. (ref. 11) (if assumed to be pinned at the Fermi Level) may correspond to the chromium impurity level at 0.73 eV below the conduction band edge, and the

states observed in the range of 0.9-1.0 eV may correspond to iron and/or copper impurities. Also, if the assumption is made that deep impurity traps cause the depletion surface region and are pinned at the Fermi level at the surface, then the data of Flinn (ref. 17) and Flinn and Briggs (ref. 18) will yield levels corresponding closely to chromium levels in n-type GaAs and oxygen levels in p-type GaAs.

The important consequence of the validity of the impurity model is that the efficiency of GaAs solar cells becomes directly linked to the purity of the host material. Improving the purity of GaAs or at least eliminating the impurities that result in deep levels will increase the efficiency for GaAs solar cells both by decreasing the surface state density and therefore lowering the SRV and also by decreasing the bulk density of traps and therefore increasing the lifetime of photo-induced charge carriers.

#### VIII. CONCLUDING REMARKS

The experimental and theoretical basis for surface photovoltage spectroscopy has been outlined. The results both reported in the literature and obtained at Langley Research Center have been reviewed and discussed. It is speculated that the surface states reported in the literature are due to deep lying bulk impurity acceptor states pinned at the Fermi level in n-type GaAs and donor states similarly pinned in p-type GaAs. These impurities could be chromium in the case of n-type and oxygen in the case of p-type GaAs. Verification of the proposed model will indicate the direction to proceed to obtain increased efficiency solar cell material.

## IX. APPENDIX

This appendix gives a calculation of the SRV using the measured parameters of the surface states on a GaAs sample as reported in reference 11 and equation (5.83) on page 197 of reference 2. The surface recombination velocity,  $S$ , is written for n-type material as

$$\frac{S}{S_{\max}} = \frac{\cosh[(E_t - E_i)/kT - U_o] + 1}{\cosh[(E_t - E_i)/kT - U_o] + \cosh(U_s - U_o)}$$

where

$$S_{\max} = \frac{\sqrt{K_p K_n} N_t n_B}{2n_i \{ \cosh[(E_t - E_i)/kT - U_o] + 1 \}}$$

$$U_o = \frac{1}{2} \ln \left( \frac{K_p}{K_n} \right)$$

where  $n_i$  and  $E_i$  are the intrinsic carrier concentration and intrinsic Fermi level;  $U_s$  is the surface potential in the dark in units of  $kT/q$ ;  $K_p$  is the capture cross section for holes multiplied by the thermal velocity; and the remaining parameters have been defined in the text. It is assumed that flat band conditions prevail (i.e., intense white light generation of carriers) and that  $U_s = U_o$  (which gives an upper limit to the SRV).  $U_s$  is related to the surface voltage (see ref. 2).

Under dark conditions for the data considered

$$U_s = 0.38$$

and it follows that

$$U_o = U_s = 1/2 \ln(K_p/K_n)$$

Using the lower limit of the capture cross section for electrons as quoted in reference 11, it is found that

$$K_n \approx 10^{-6} \text{ cm}^3/\text{sec}$$



at room temperature, and

$$K_p/K_n \approx 2.1.$$

Then

$$S_{\max} = 8 \times 10^{14} \text{ cm/sec.}$$

For flat band conditions, the SRV is

$$S \approx S_{\max} \times 2.7 \times 10^{-9}$$

or

$$S \approx 2.2 \times 10^6 \text{ cm/sec.}$$

This result assumed a bulk concentration of majority carriers to be  $4 \times 10^{14}/\text{cm}^3$ . If the same parameters are used for a bulk concentration of majority carriers of  $10^{17}/\text{cm}^3$ , then the SRV has a value of

$$S \approx 3 \times 10^5 \text{ cm/sec.}$$

Both of these values are in good agreement with measured values (refs. 17 and 18) for the SRV on GaAs which range from  $1.34 \times 10^4$  to  $1.1 \times 10^6$  cm/sec.

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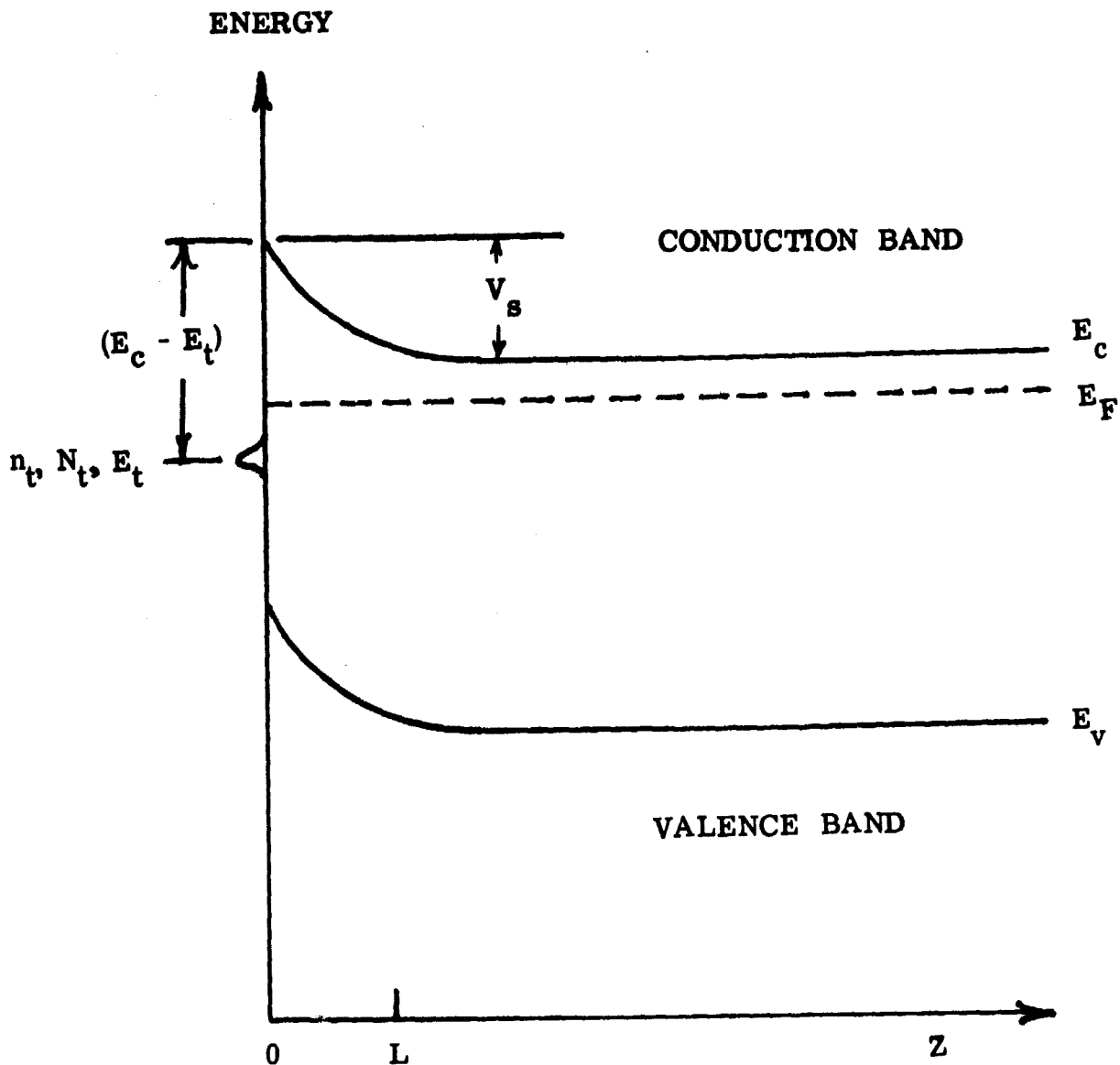


Figure 1. Schematic of the surface region of an n-type semiconductor for the case of a depletion layer where  $E_C$  is the energy of the conduction band edge,  $E_F$  is the Fermi energy, and  $E_V$  is the energy of the valence band edge.

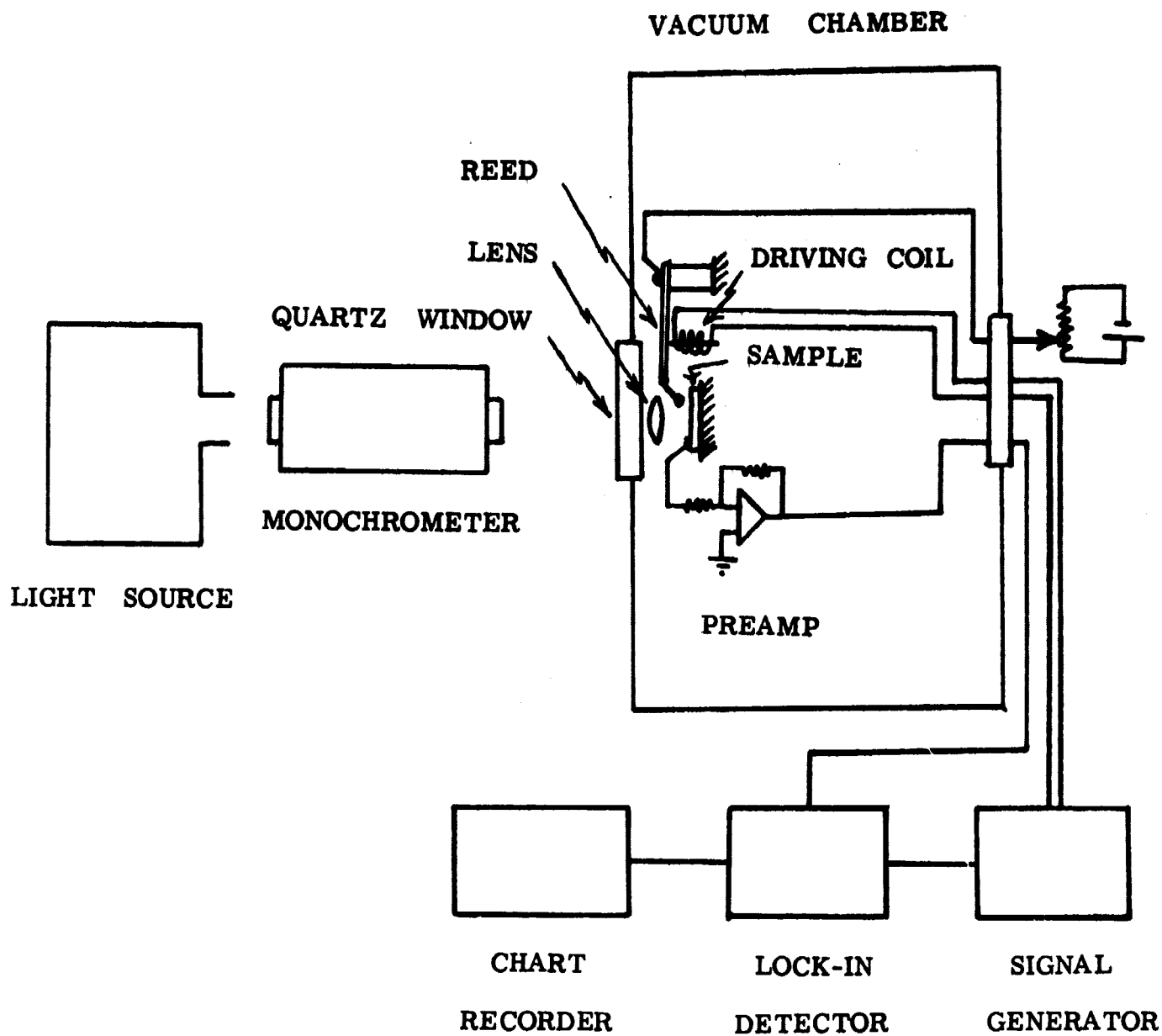


Figure 2. Schematic diagram of the experimental set-up.

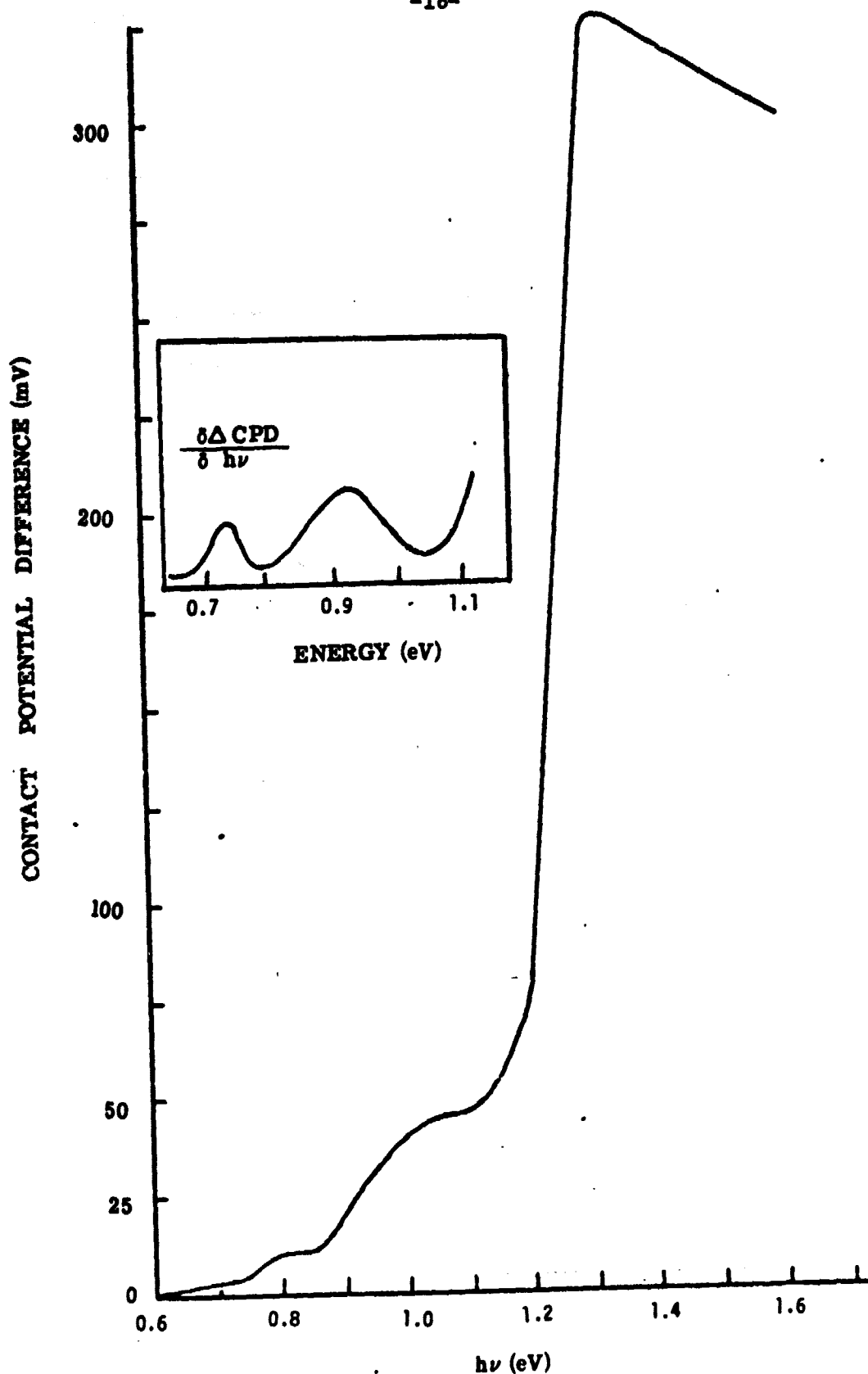


Figure 3. Photovoltage spectrum of the (111) surface of GaAs (from ref. 11).

Insert shows the derivative of the photovoltage curve with respect to photon energy.

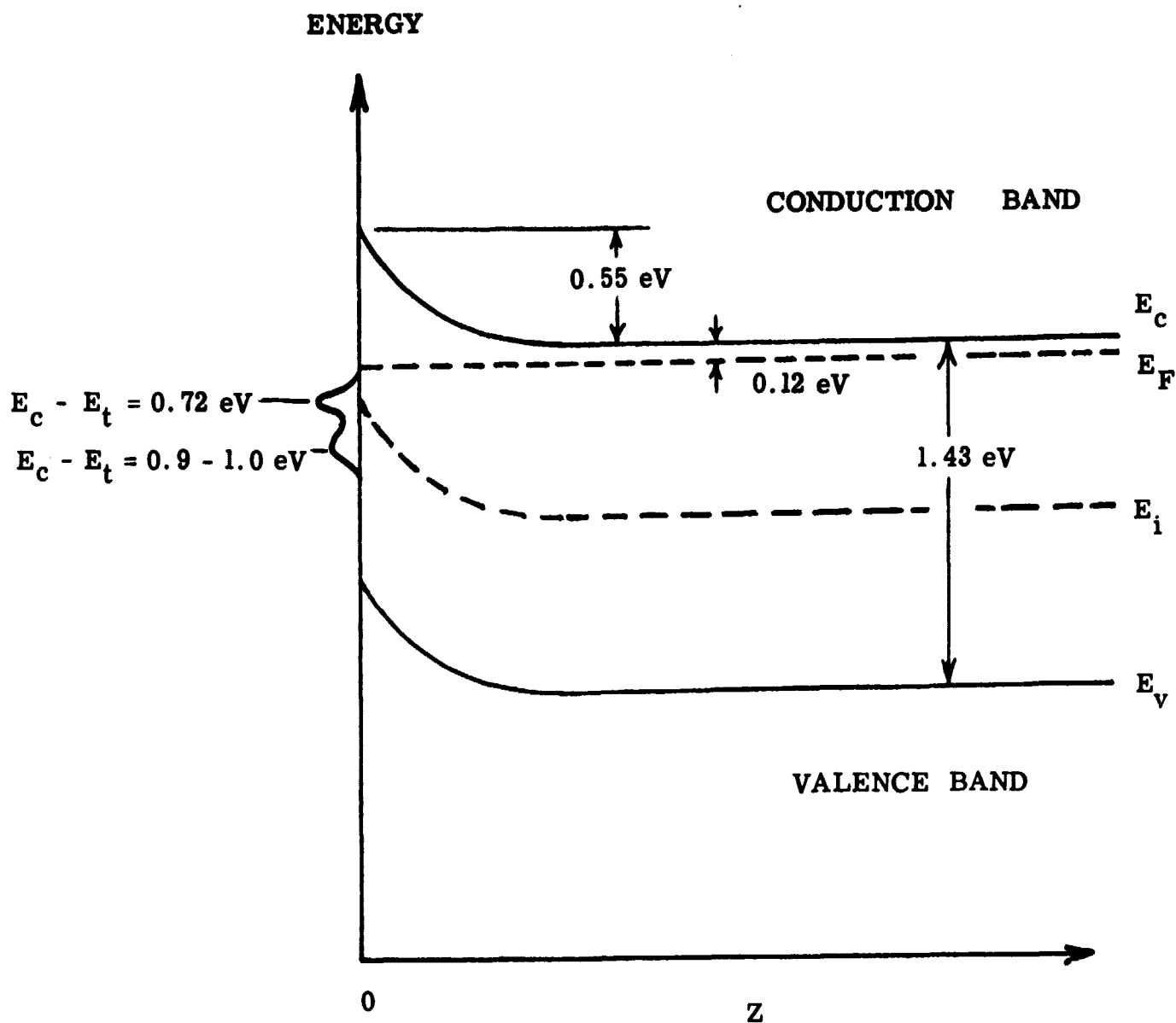


Figure 4. Schematic representation of the surface states indicated by the data shown in fig. 3, where  $E_C$  is the energy of the conduction band edge,  $E_F$  is the Fermi energy,  $E_i$  is the intrinsic energy, and  $E_V$  is the energy of the valence band edge.

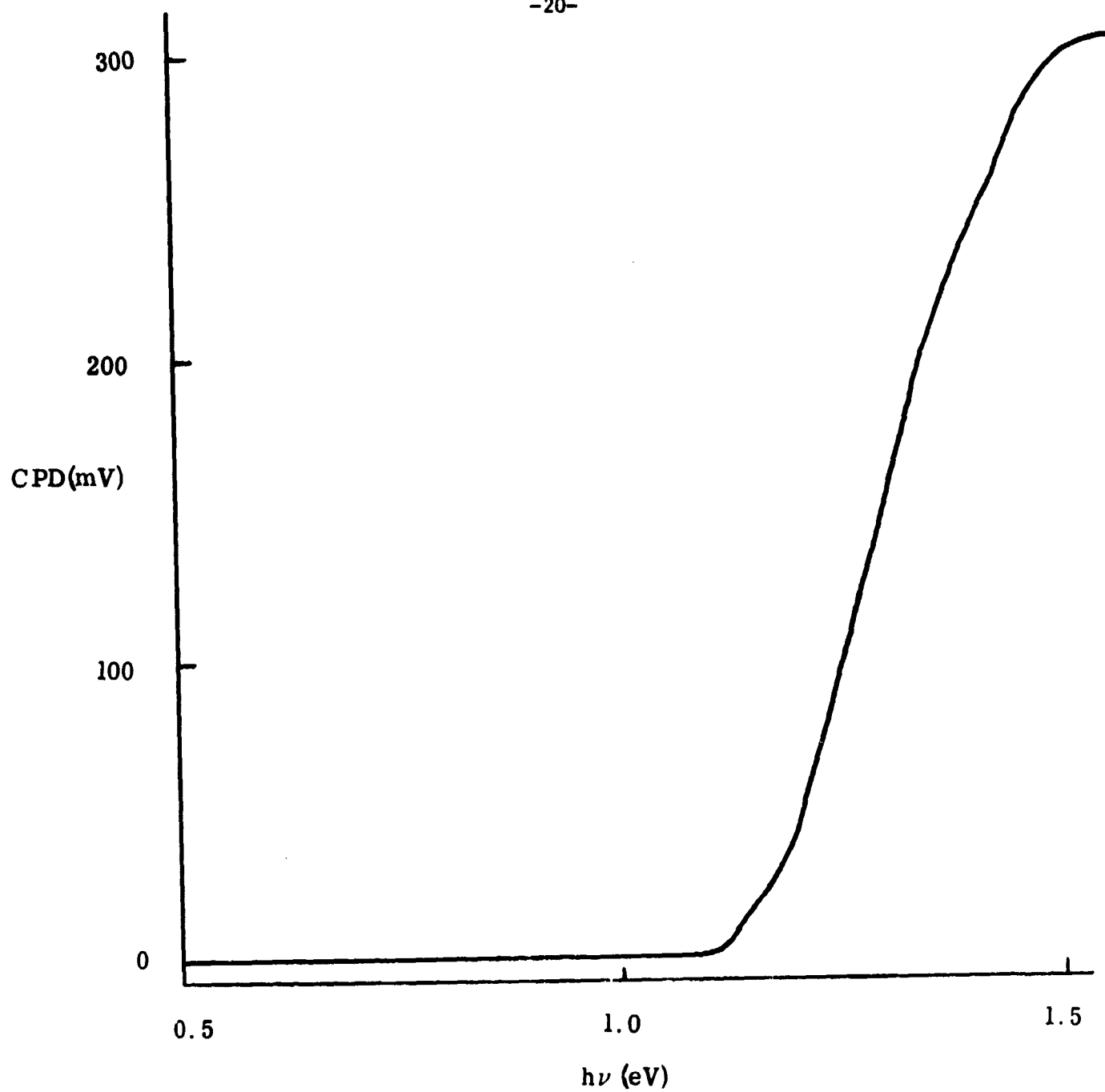


Figure 5. Photovoltage spectrum of (100) surface of n-type GaAs in vacuum.